

1,075,166



PATENT SPECIFICATION

NO DRAWINGS

1,075,166

Inventor: WILLIAM CUMMINGS

Date of filing Complete Specification: April 26, 1966.

Application Date: June 1, 1965. No. 23261/65.

Complete Specification Published: July 12, 1967.

© Crown Copyright 1967.

Index at Acceptance:—C2 C(1Q6B1, 1Q6B2, 1Q8A, 1Q9A, 1Q11G)

Int. Cl.:—C 07 e 103 / 30

COMPLETE SPECIFICATION

Fluorinated Aromatic Diamides

We, MONSANTO CHEMICALS LIMITED, a British Company of Monsanto House, 10—18 Victoria Street, London, S.W.1, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to new chemical compounds, in particular to new fluorine-containing aromatic amides.

The new compounds are diamides having the formula $A-Ar-A$ wherein Ar represents a divalent aromatic structure containing one or more aromatic nuclei, A represents an amide grouping having the formula $R.CO.N(R')$ — or $R.N(R').CO$ — attached to a nuclear carbon atom of the aromatic structure Ar , R represents a monovalent aromatic radical containing one or more aromatic nuclei, R' represents a hydrogen atom or a monovalent aromatic radical containing one or more aromatic nuclei, and where there is present at least one fluorine atom as a nuclear substituent in the diamide molecule.

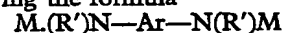
The compounds have good thermal stability and are useful as components in compositions that are exposed to high working temperatures.

The invention includes a process for the production of a new diamide of the invention, in which a diamine having the formula $R'NH-Ar-NHR'$ or a mono-amine having the formula $R.R'NH$ is reacted respectively with a monocarboxylic halide having the formula $R.CO.X$ or a dicarboxylic halide having the formula $X.CO.Ar.CO.X$, where X represents a halogen atom and Ar , R and R' have the same significance as in the formula of the product, provided that where R' represents an aromatic radical at least one of the aromatic nuclei to which each nitrogen atom is linked is free from fluorine.

The process is preferably conducted in solution in the presence of an acid-binding agent.

Also part of the invention is a process for the production of a new diamide of the invention

in which both R and R' represent monovalent aromatic radicals, which comprises the reaction of an alkali metal derivative of a di-secondary amine having the formula



or an alkali metal derivative of a mono-secondary amine having the formula $RR'NM$, where in each instance M represents an alkali metal atom, with respectively a monocarboxylic halide having the formula $R.CO.X$ or a dicarboxylic halide having the formula $X.CO.Ar.CO.X$ where X represents a halogen atom and Ar , R and R' have the same significance as in the formula of the product.

The process is preferably conducted in an inert liquid medium at ordinary room temperatures or below.

In the new diamides, fluorine may be present in any one or more of Ar , R or R' , and where fluorine is present, the preferred structures and radicals are those that are fully fluorinated. R , for instance, can be a phenyl or fluorinated phenyl radical, and where it is a fluorinated phenyl radical, it is preferably a pentafluorophenyl radical.

In respect of the divalent aromatic structure Ar , the diamides that are most readily obtainable are usually those where Ar represents a phenylene, fluorinated phenylene, biphenylene, or fluorinated biphenylene group; in respect of the monovalent radical R , the most readily obtainable diamides are usually those where R represents a phenyl, fluorinated phenyl, biphenyl or fluorinated biphenyl group; and in respect of the radical R' , the most readily obtainable diamides are usually those where R' represents a hydrogen atom or a phenyl, fluorinated phenyl, biphenyl or a fluorinated biphenyl radical.

More generally however, the class of new diamides includes members where Ar represents for instance an optionally fluorinated naphthalene nucleus, or two optionally fluorinated benzene nuclei linked to each other through an intermediate atom or group, for

[Price 4s. 6d.]

example an oxygen or sulphur atom, or an alkylene, carbonyl or sulphonyl group.

- Similarly in the general class of diamides, R or R' can be for instance a naphthyl radical, a radical comprising two benzene nuclei linked to each other through an intermediate atom or group, for instance an oxygen or sulphur atom, or an alkylene, carbonyl or sulphonyl group, or an analogous fluorine-containing radical.
- Specific examples of new diamides of the invention are: 1,3-bis(pentafluorobenzamido) benzene; 1,4-bis(pentafluorobenzamido) benzene; 1,3-bis(pentafluorobenzamido) tetrafluorobenzene; 1,3-bis[p-(pentafluorophenyl) tetrafluorobenzamido] benzene; N,N'-bis(pentafluorophenyl)-1,4-bis(pentafluorobenzamido) tetrafluorobenzene; N,N'-bis(pentafluorophenyl)-1,4-di(benzamido) tetrafluorobenzene; N,N'-diphenyl-1,4-bis(pentafluorobenzamido)benzene; 4,4'-bis(benzamido)-2,2',3,3',5,5',6,6'-octafluorobiphenyl; 4,4'-bis(pentafluorobenzamido) biphenyl; 4,4'-bis(pentafluorobenzamido)-2,2',3,3',5,5',6,6'-octafluorobiphenyl; 2,6-bis(pentafluorobenzamido) naphthalene; 4,4'-bis(pentafluorobenzamido)octafluorodiphenyl ether; 4,4'-bis[p-(pentafluorophenyl) tetrafluorobenzamido] methane; 1,4-bis[N(pentafluorophenyl) benzamido] tetrafluorobenzene; isophthaloyl di(pentafluoroanilide); tetrafluoroisophthaloyl di(pentafluoroanilide); terephthaloyl di(pentafluoroanilide); tetrafluoroterephthaloyl dianilide; tetrafluoroterephthaloyl di(pentafluoroanilide); N,N,N',N'-tetra(pentafluorophenyl) tetrafluoroterephthalamide; 2,7-bis(pentafluorophenylaminocarbonyl) naphthalene (pentafluorophenylaminocarbonyl)naphthalene; 4,4'-bis(pentafluorophenylaminocarbonyl) biphenyl; 3,3'-bis(pentafluorophenylaminocarbonyl) octafluorobiphenyl; 2,4'-bis(pentafluorophenylaminocarbonyl) diphenyl ether; 2,2'-bis[p(pentafluorophenylaminocarbonyl) tetrafluorophenyl] propane; and 4,4'-bis(pentafluorophenylaminocarbonyl) diphenyl sulphone.

- The preferred carboxylic halides for use in the processes of the invention are the chlorides, although the fluorides, bromides and iodides are also operative.

- With regard to the aromatic structure Ar and the radicals R and R', it is readily apparent which reactants are required for the production of any particular diamide. Where the process comprises the reaction of a diamine with a monocarboxylic halide having the formula $R.CO.X$, representative di-primary amines of the formula $H_2N-Ar-NH_2$ are o-phenylenediamine; p-phenylenediamine; tetrafluoro-m-phenylenediamine; 4,4'-diaminobiphenyl; 3,3'-diamino-octafluorobiphenyl; 4,4'-diamino-octafluorobiphenyl; 2,7-diaminonaphthalene; and 4,4'-diamino-octafluorodiphenyl ether; representative di-secondary amines are N,N-diphenyl-p-phenylenediamine; N,N'-di(pentafluorophenyl)-p-phenylenediamine; and N,N'-di-(pentafluorophenyl)-tetrafluoro-m-phenylenediamine (at least the last-named being em-

ployed in the form of its alkali metal derivative); and representative monocarboxylic halides are benzoyl chloride, pentafluorobenzoyl chloride; 4(pentafluorophenyl) tetrafluorobenzoyl chloride; and 2-naphthoyl chloride.

Where the process comprises the reaction of a mono-amine or an alkali metal derivative of a mono-secondary amine with a dicarboxylic halide, representative amines are aniline, pentafluoroaniline, 4-aminobiphenyl; 4-aminononafluorobiphenyl; 3-aminodiphenyl sulphone; 2,3,4,5,6-pentafluorodiphenylamine; and decafluorodiphenylamine; and representative dicarboxylic halides are phthaloyl chloride; isophthaloyl chloride; tetrafluoroisophthaloyl chloride; terephthaloyl chloride; tetrafluoroisophthaloyl chloride; naphthoyl chloride; 2,3,5,6,7,8-hexafluoronaphthalene-1,4-dicarboxylic chloride; biphenyl 4,4'-dicarboxylic chloride and octafluorobiphenyl-4'4'-dicarboxylic chloride.

Where the process in which a primary amine is a reactant is conducted in the presence of an acid-binding agent, this preferably a tertiary amine, for example tributylamine, dimethylaniline, diethylaniline, pyridine or quinoline, but inorganic compounds, such as for instance the alkali metal carbonates and bicarbonates, and the oxides, hydroxides and carbonates of the alkaline earth metals, can be used. The quantity of the acid-binding agent used is normally at least sufficient to neutralise the theoretical quantity of hydrogen halide eliminated.

Suitable solvents for use in the process include hydrocarbons, especially aromatic hydrocarbons, for instance benzene and toluene, halogenated hydrocarbons, for instance chloroform and perchlorethylene, and ethers, for instance diethyl ether, dioxan, and tetrahydrofuran. Certain tertiary amines can function both as acid-binding agent and as solvent.

Reaction temperatures in this process are not critical; the reaction between the primary amine and the acid halide proceeds on mixing the reactants at ordinary temperatures. It is usually preferred however to maintain the reaction mixture at a somewhat elevated temperature, for example at a temperature in the range from 50° to 150°C at least for a time after mixing is complete, in order to ensure completion of the reaction.

In the process in which an alkali metal derivative of a secondary amine is employed, the alkali metal derivative can be for example a lithium, sodium or potassium derivative and can be obtained from the appropriate amine in a generally conventional manner, for example by the action of an alkali metal alkyl, hydride or alkoxide. For the production of a lithium derivative, the lithium alkyls are often the most satisfactory reagents, and operation at low temperatures, for example in the range of -20° to -100°C. is desirable for maximum yields. Sodium and potassium derivatives can conveniently be obtained from the correspond-

ing hydrides, and potassium derivatives can be prepared from potassium alkoxides, for example potassium ethoxide. Operation in an inert atmosphere is again desirable, but these processes can be conducted at ordinary or slightly elevated temperatures, for example at 5° to 45°C. Heating to temperatures in excess of, say, 60°C. is best avoided, however.

Ethers, for example diethyl ether or tetrahydrofuran, are suitable liquid media for the reaction of an alkali metal derivative with a carboxylic halide. Reaction temperatures not exceeding -20°C. where the alkali metal derivatives is a lithium derivative, and not exceeding 60°C. and more especially within the range 5 to 45°C. where the alkali metal derivative is a sodium or potassium derivative, are generally preferred.

Each instance of the process involves the reaction of a difunctional reactant with a monofunctional reactant, and usually there are employed approximately 2 mols of the monofunctional reactant per mol of the difunctional reactant. The use of an excess, for example up to 10% excess, of the monofunctional reactant can lead to higher yields of the product in some instances.

The invention is illustrated by the following Examples.

Example 1

This Example describes the production of the new compound 1,3-bis(pentafluorobenzamido) benzene.

A solution of 6.92 grams (0.03 mol.) of pentafluorobenzoyl chloride in 10 cc. of benzene was added to a stirred solution of 1.62 grams (0.015 mol.) of *m*-phenylenediamine and 5.5 cc. of *N,N*-diethylaniline in 40 cc. of benzene, and the reaction mixture was then boiled under reflux for 5 hours. After cooling, the mixture was stirred at room temperature with an equal volume of *N* hydrochloric acid, the product was filtered, washed with water and dried. Crystallisation from aqueous ethanol gave 7.0 grams of 1,3-bis(pentafluorobenzamido) benzene as colourless prisms having a melting point of 260—261°C. (Found: C, 48.8; H, 0.9; F, 38.2; N, 5.8%. $C_{20}H_8F_{10}N_2O_2$ requires C, 48.4; H, 1.2; F, 38.3; N, 5.65%).

Example 2

This Example describes the production of the new compound 1,4-bis(pentafluorobenzamido) tetrafluorobenzene.

A solution of 4.75 grams of pentafluorobenzoyl chloride, 1.85 grams of tetrafluoro-*p*-phenylenediamine and 3.5 cc. of *N,N*-diethylaniline in 40 cc. of benzene was boiled under reflux with stirring for 5 hours. After cooling, the reaction mixture was shaken with 25 cc. of 2*N* hydrochloric acid. The solid product was collected by filtration and washed with water and acetone. Crystallisation from a mixture of ethanol and *N,N*-dimethylformamide gave 3.7 grams of 1,4-bis(pentafluorobenzamido) tetrafluorobenzene having a melt-

ing point of 353.5—355.5°C. (decomp.) (Found: C, 42.3; H, 0.1; F, 46.9; N, 5.0. $C_{20}H_2F_{14}N_2O_2$ requires: C, 42.3; H, 0.35; F, 46.8; N, 4.95%).

Example 3

Using essentially the same procedure as that described in Example 1, 4.5 grams of 4,4'-dibenzamidooctafluorobiphenyl having a melting point of 363—365°C. (decomp.) were obtained from 4.92 grams of 4,4'-diaminooctafluorobiphenyl and 4.22 grams of benzoyl chloride, the initial product being crystallised from a mixture of 2-ethoxyethanol and *N*-methyl-2-pyrrolidone. (Found: C, 57.9; H, 2.35; F, 26.1; N, 5.4%. $C_{26}H_{12}F_8N_2O_2$ requires C, 58.2; H, 2.25; F, 28.35; N, 5.2%).

Example 4

Using essentially the same procedure as that described in Example 1, 7.5 grams of 4,4'-bis(pentafluorobenzamido) biphenyl having a melting point of 350—352°C. (decomp.) were obtained from 2.76 grams of 4,4'-diaminobiphenyl and 6.92 grams of pentafluorobenzoyl chloride, the initial product being crystallised from a mixture of 2-ethoxyethanol and *N*-methyl-2-pyrrolidone. (Found: C, 54.7; H, 1.8; F, 33.0; N, 4.9%. $C_{26}H_{10}F_{10}N_2O_2$ requires C, 54.55; H, 1.75; F, 33.2; N, 4.9%).

Example 5

Using essentially the same procedure as that described in Example 1, 4.5 grams of 4,4'-bis(pentafluorobenzamido) octafluorobiphenyl having a melting point of 359—361°C. were obtained from 4.92 grams of 4,4'-diaminooctafluorobiphenyl and 6.92 grams of pentafluorobenzoyl chloride, the initial product being crystallised from a mixture of 2-ethoxyethanol and *N*-methyl-2-pyrrolidone (Found: C, 43.95; H, 0.33; F, 46.15; N, 4.1%. $C_{26}H_8F_{18}N_2O_2$ requires C, 43.6; H, 0.28; F, 47.75; N, 3.9%).

Example 6

This Example describes the production of the new compound isophthaloyl di(pentafluoroanilide).

A Mixture of 7.32 grams (0.04 mol.) of pentafluoroaniline 4.06 grams (0.02 mol.) of isophthaloyl chloride and 6.55 grams (0.044 mol.) of *N,N*-diethylaniline in 50 cc. of benzene was boiled for 5 hours with stirring under reflux.

After cooling, the mixture was stirred at room temperature for 30 minutes with an equal volume of *N* hydrochloric acid, and the solid product was collected by filtration and washed with water. Crystallisation from aqueous ethanol and recrystallisation from acetic acid gave 9.5 grams of isophthaloyl di(pentafluoroanilide) as colourless needles having a melting point of 273—275°C. (Found: C, 48.65; H, 1.25; F, 38.15; N, 5.8%. $C_{20}H_6F_{10}N_2O_2$ requires C, 48.4; H, 1.2; F, 38.3; N, 5.65%).

Example 7

Using essentially the same procedure as that described in Example 6, 9.5 grams of terephthaloyl di(pentafluoroanilide) having a melting point of 328—330°C. (decomp.) were obtained

by the reaction of 7.32 grams of pentafluoroaniline and 4.06 grams of terephthaloyl chloride in the presence of N,N-diethylaniline, the initial product being crystallised from a mixture of 2-ethoxyethanol and N-methyl-2-pyrrolidone. (Found: C, 48.4; H, 1.6; F, 38.15; N, 5.75%. $C_{20}H_6F_{10}N_2O_2$ requires C, 48.4; H, 1.2; F, 38.3; N, 5.65%.)

Example 8

This Example describes the production of the new compound tetrafluoroterephthaloyl dianilide.

A solution of 5.6 gram (0.02 mol.) of tetrafluoroterephthaloyl chloride in 30 cc. of pyridine was added to a stirred solution of 3.9 grams (0.042 mol.) of aniline in 20 cc. of pyridine, initially at room temperature. The reaction mixture became warm during the addition and was afterwards heated to boiling under reflux for 10 minutes. After cooling, the reaction mixture was poured into 400 cc. of water, whereupon tetrafluoroterephthaloyl dianilide separated as an insoluble solid in a yield of 7.5 grams. The dianilide was obtained in the form of colourless plates having a melting point of 314–317°C. (decomp.) by crystallisation from a mixture of ethanol and N-methyl-2-pyrrolidone and recrystallisation from acetic acid. (Found: C, 62.15; H, 3.4; N, 7.35%. $C_{20}H_{12}F_4N_2O_2$ requires C, 61.85; H, 3.1; N, 7.2.)

Example 9

Using essentially the same procedure as that described in Example 6, 10.5 grams of tetrafluoroterephthaloyl bis(pentafluoroanilide) were obtained by the reaction of 7.32 grams of pentafluoroaniline with 5.5 grams of tetrafluoroterephthaloyl chloride in the presence of N,N-diethylaniline. The product was crystallised from a mixture of 2-ethoxyethanol and N-methyl-2-pyrrolidone giving colourless needles having a melting point of 350–352°C. (Found: C, 42.05; H, 0.9; F, 46.25; N, 4.45, 5.4. $C_{20}H_2F_{14}N_2O_2$ requires: C, 42.3; H, 0.35; F, 46.8; N, 4.95%.)

Example 10

This Example describes the production of the new compound tetrafluoroisophthaloyl di(pentafluoroanilide).

A mixture of 7.32 grams of pentafluoroaniline 5.5 grams of tetrafluoroisophthaloyl chloride and 7 cc. of N,N-diethylaniline in 60 cc. of benzene was boiled under reflux with stirring for 5 hours. After cooling, the reaction mixture was shaken with 50 cc. of N-hydrochloric acid, and the solid which precipitated was collected and washed with water and with benzene. Recrystallisation from aqueous ethanol gave 9 grams of tetrafluoroisophthaloyl di(pentafluoroanilide) as colourless prisms having a melting point of 323–325°C. (decomp.) (Found: C, 42.2; H, 0.5; F, 46.95; N, 5.0%. $C_{20}H_2F_{14}N_2O_2$ requires C, 42.3; H, 0.35; F, 46.8; N, 4.95%.)

Example 11

This Example describes the production of the new compound N,N,N',N'-tetra(pentafluorophenyl) tetrafluoroterephthalamide.

A solution of 2.4 cc. of n-butyl bromide in 40 cc. of ether was added during 45 minutes to a stirred suspension of 1 gram of finely-divided lithium in 65 cc. of ether at 5–10°C. in an atmosphere of nitrogen. The mixture was stirred at 5°C. for a further 30 minutes and was then allowed to warm up to room temperature during 1½ hours. The solution of butyl lithium thus obtained was separated from unreacted lithium by filtration through glass wool.

The solution was then cooled to –60°C., and, in an atmosphere of nitrogen, a solution of 6.98 grams of decafluorodiphenylamine in 50 cc. of tetrahydrofuran was added with stirring during 30 minutes, the temperature being maintained at –60°C. After a further 30 minutes a solution of 2.75 grams of tetrafluoroterephthaloyl chloride in 60 cc. of tetrahydrofuran, was added with stirring at –60°C. during 15 minutes. After a further 15 minutes at –60°C., the reaction mixture was allowed to warm to room temperature and was then stirred with 3 cc. of water and 1 cc. of 2N hydrochloric acid. The product, which was insoluble in the reaction mixture, was collected by filtration and washed with ether. The yield was 8 grams. A 7 gram portion was crystallised from N,N-dimethylformamide and recrystallised successively from xylene and methyl ethyl ketone to give 2.1 grams of N,N,N',N'-tetra(pentafluorophenyl) tetrafluoroterephthalamide as colourless prisms having a melting point of 291–292°C. (Found: C, 42.35; H, 0.15; N, 3.2%. $C_{32}F_{24}N_2O_2$ requires C, 42.7; H, 0.0; N, 3.1%.)

Example 12

This Example describes the production of N,N,N',N'-tetra (pentafluorophenyl) tetrafluoroterephthalamide via the potassium derivative of decafluorodiphenylamine.

A solution of 0.272 gram (4.85 millimol) of potassium hydroxide in 3 cc. of ethanol was added to a solution of 1.694 gram (4.85 millimol) of decafluorodiphenylamine in 4 cc. of ethanol. The ethanol was then evaporated at room temperature under reduced pressure giving the required potassium derivative as a pale greenish-yellow solid. This was next dissolved in 40 cc. of ether, and to the solution then was added 0.632 gram (2.3 millimol) of tetrafluoroterephthaloyl chloride. After stirring for 2 hours at room temperature (20°C.) and standing overnight, the mixture was filtered. The solid thus collected was washed with dilute hydrochloric acid, with water until the washing was free from acid, and was then dried to give 1.36 grams (62% yield) of crude N,N,N',N'-tetra(pentafluorophenyl) tetrafluoroterephthalamide having a melting point of about 280°C. This was raised to 289–291°C. by crystallisation from xylene, and was unde-

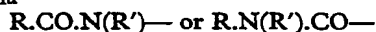
pressed by admixture with a sample of the N,N,N',N'-tetra (pentafluorophenyl) tetrafluoroterephthalamide prepared in the manner described in Example 11.

5 Example 13

This Example describes the production of the new compound N,N'-bis (pentafluorophenyl)-1,4-bis(pentafluorobenzamido) tetrafluorobenzene. A solution of 5.38 grams of N,N'-bis(pentafluorophenyl) tetrafluoro-p-phenylenediamine in 60 cc. of ether was added to a suspension of 0.504 gram of sodium hydride in 75 cc. of ether prepared under nitrogen. After refluxing for 2 hours, 4.48 grams of pentafluorobenzoyl chloride were added to the reaction mixture and refluxing was continued for a further 2 hours. The product was filtered, and the solid collected was washed with dilute hydrochloric acid and then with water until free from acid to yield, after drying, 8.4 grams of N,N'-bis(pentafluorophenyl)-1,4-bis (pentafluorobenzamido) tetrafluorobenzene having a melting point of 282—3°C. The diamide was recrystallised from aqueous dimethylacetamide and from methyl ethyl ketone, but the melting point remained constant.

WHAT WE CLAIM IS:—

1. A diamide having the formula A—Ar—A where Ar represents a divalent aromatic structure containing one or more aromatic nuclei, A represents an amide grouping having the formula



attached to a nuclear carbon atom of the aromatic structure Ar, R represents a monovalent aromatic radical containing one or more aromatic nuclei, R' represents a hydrogen atom or a monovalent aromatic radical containing one or more aromatic nuclei, and where there is present at least one fluorine atom as a nuclear substituent in the diamide molecule.

2. A diamide according to Claim 1, in which any aromatic nucleus that contains fluorine is fully fluorinated.

3. A diamide according to either of Claims 1 and 2, in which Ar represents a phenylene, fluorinated phenylene, biphenylene or fluorinated biphenylene group.

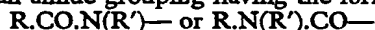
4. A diamide according to any of Claims 1 to 3, in which R represents a phenyl, fluorinated phenyl, biphenyl or fluorinated biphenyl group.

5. A diamide according to any of Claims 1 to 4, in which R' represents a hydrogen atom or a phenyl or fluorinated phenyl radical.

6. A diamide according to Claim 1 substantially as described in any of Examples 1 to 11.

7. A diamide according to Claim 1 substantially as described in Example 13.

8. A process for the production of a diamide having the formula A—Ar—A wherein Ar represents a divalent aromatic structure containing one or more aromatic nuclei, A represents an amide grouping having the formula



attached to a nuclear carbon atom of the aromatic structure Ar, R represents a monovalent aromatic radical having one or more aromatic nuclei, R' represents a hydrogen atom or a monovalent aromatic radical having one or more aromatic nuclei and where Ar, R and R' together contain at least one fluorine atom as a nuclear substituent, provided that where A represents the grouping R.CO.N(R')— and R' is an aromatic radical, at least one of Ar and R' is free from fluorine, and that where A represents the grouping R.N(R').CO— and R' is an aromatic radical at least one of the aromatic nuclei to which each nitrogen atom is linked is free from fluorine, in which a diamine having the formula R'NH—Ar—NHR' or a mono-amine having the formula RR'NH is reacted respectively with a monocarboxylic halide having the formula R.CO.X or a dicarboxylic halide having the formula X.CO.Ar.CO.X where X represents a halogen atom and Ar, R and R' have the same significance as in the formula of the product.

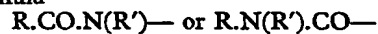
9. A process according to Claim 8 that is conducted in solution in the presence of an acid-binding agent.

10. A process according to either of Claims 8 and 9 for the production of a diamide characterised by the features specified in any of Claims 2 to 5.

11. A process according to Claim 8 substantially as described in any of Examples 1 to 10.

12. A diamide that has been produced by a process according to any of Claims 8 to 11.

13. A process for the production of a diamide having the formula A—Ar—A wherein Ar represents a divalent aromatic structure containing one or more aromatic nuclei, A represents an amide grouping having the formula



attached to a nuclear carbon atom of the aromatic structure Ar, each of R and R' represents a monovalent aromatic radical having one or more aromatic nuclei, and where Ar, R and R' together contain at least one fluorine atom as a nuclear substituent, which comprises the reaction of an alkali metal derivative of a di-secondary amine having the formula M.(R')N—Ar—N(R').M or an alkali metal derivative of a mono-secondary amine having the formula RR'NM, where in each instance M represents an alkali metal atom, with respectively a monocarboxylic halide having the formula R.CO.X or a dicarboxylic halide having the formula X.CO.Ar.CO.X where X represents a halogen atom and Ar, R and R' have the same significance as in the formula of the product.

14. A process according to Claim 13, in which the alkali metal derivative is a lithium derivative and the reaction is carried out in an inert liquid medium at a temperature within the range —20° to —100°C.

15. A process according to either of Claims 130

- 13 and 14 for the production of a diamide characterised by the features specified in any of Claims 2 to 5.
16. A process according to Claim 13 substantially as described in Example 11. 5
17. A diamide that has been obtained by a process according to any of Claims 13 to 16.
18. A process according to Claim 13, in which the alkali metal derivative is a sodium or potassium derivative and the process is carried out in an inert liquid medium at a temperature within the range 5 to 45°C. 10
19. A process according to Claim 18 for the production of a diamide characterised by the features specified in any of Claims 2 to 5. 15
20. A process according to Claim 18 substantially as described in either of Example 12 and 13.
21. A diamide that has been obtained by a process according to any of Claims 18 to 20. 20
- C. G. WICKHAM,
Chartered Patent Agent,
Monsanto House, 10-18 Victoria Street,
London, S.W.1.

Printed for Her Majesty's Stationery Office by J. Looker Ltd., Poole, Dorset. 1967.
Published at The Patent Office, 25, Southampton Buildings, London, W.C.2, from which copies may be obtained.